Halocarbons in the TTL: the roles of oceanic emissions and atmospheric transport

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Global modelling studies of brominated and iodinated halocarbons often rely on annual mean emission scenarios following the top-down approach. One drawback of this approach is the adjustment of the emissions in order to reproduce upper-air observations without taking into account seasonal or interannual variations in either of the two factors. Recent biogeochemistry-halocarbon modelling has shown, however, that temporal variations in the oceanic production can drive a strong seasonal reversal of the emissions. Comprehensive methods combining our understanding of oceanic and atmospheric processes are required in order to derive reliable estimates of halocarbon emissions and TTL abundances.

In this study, we will use monthly mean emissions of brominated and iodinated halocarbons to derive their contribution to the global stratospheric halogen loading based on transport calculations with the Lagrangian particle dispersion model FLEXPART. We will analyze how seasonality in both processes, marine emissions and atmospheric transport, drives the seasonality of the halocarbon distribution in the TTL. Aircraft observations from the last 25 years are classified according to the temporal variability of the halocarbon distribution derived from the FLEXPART simulations. Thus, we are able to identify which aircraft observations can be considered representative for the mean halocarbon abundance and which observations are strongly tied to the seasonal variations in emissions and transport. In combination with backward trajectory runs from the aircraft locations to the surface, we will show where upper-air observations are consistent with our understanding of emissions and transport and where marine sources are missing. Based on the global Lagrangian transport calculations, the overall contributions of various oceanic regions to the TTL halogen budget are determined. In addition, the importance of halogen emissions from the open ocean versus emissions from coastal regions for the TTL budget is discussed. By combining up-to-date emission scenarios, global Lagrangian transport calculations and existing aircraft measurements our study identifies where and when future oceanic and upper-air measurements are necessary to enhance our understanding of marine halocarbons in the TTL.